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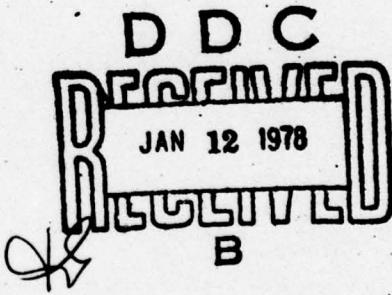
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CHARGE TRANSFER PUMPING OF THE HELIUM-NITROGEN LASER AT ATMOSPHERIC PRESSURES IN AN ELECTRICAL AVALANCHE DISCHARGE

by

C. B. COLLINS, J. M. CARROLL, and K. N. TAYLOR



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21. ABSTRACT (Continue on reverse side if necessary and identify by block number) An Atmospheric Electrical Avalanche (AEA)laser, stabilized by displacement current preionization, has been developed to support the study of the collisional pumping of the N ₂ ⁴⁺ , B ₂ X, electronic transition by the kinetic step He ₂ ⁺⁺ + N ₂ + He → N ₂ ⁺ (B ₂ ^Σ) + 3He. With proper preionization, the AEA laser has been operated at a PRF of 1 to 30 Hz in an avalanche mode at 100 to 200 A/cm ² and E/p of 5 v/cm/torr. At pressures from 1 to 8 atm an essentially uniform, 30 cm ³ volume containing a high concentration of He ₂ ⁺⁺ has been produced. Resulting laser output pumped by the			

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charge transfer reaction has exceeded 1MW peak power at 427.8 nm. in a 4 nsec pulse. Efficiency with respect to the electrical power flowing through the laser tube has exceeded 2% and output pulse energies have exceeded 1% of the input pulse of energy dissipated in the laser tube.

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CHARGE TRANSFER PUMPING OF THE HELIUM-NITROGEN
LASER AT ATMOSPHERIC PRESSURES IN AN
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TECHNICAL REPORT

CHARGE TRANSFER PUMPING OF THE HELIUM-
NITROGEN LASER AT ATMOSPHERIC PRESSURES
IN AN ELECTRICAL AVALANCHE DISCHARGE

I. INTRODUCTION

In principle, charge transfer offers a considerable advantage over other laser pumping mechanisms because of the large cross-sections^{1,2} characteristic of such processes. These values lead to reaction rates which are at least an order of magnitude larger than those of other excitation transfer sequences involving neutral atomic and molecular species. As a consequence, the laser pumping reactions can be readily arranged to be the dominant processes for loss of the ionization created in a plasma. This can be done with relatively small concentrations of the gas to be excited which, in turn, means that chemical quenching of the final excited state population should be virtually negligible in comparison to stimulated emission, as seems to be the case in the helium-nitrogen laser.³ For example, as will be shown in this work, only 6 torr of N₂ represents the optimum concentration in 5 atm. of helium when excited in a preionized discharge.

Recently, new kinetic mechanisms have been described which further enhance the promise of the charge transfer laser. Multibody charge transfer reactions have been reported² which raise substantially the rates at which the principal pumping reactions can proceed. Additionally, it has been recognized⁴ that the kinetic sequence involving an ion-electron capture followed by autoionization⁵ offers a mechanism which is unique to molecular ions for the quenching of vibrational excitation. Thus,

if the lower laser level is arranged to be a vibrationally excited state of a molecular ion, the capture-autoionization process offers a rapid kinetic channel for its depopulation. These particular features of the elementary kinetic steps contribute to the following important advantages of the charge transfer scheme of laser pumping:

- 1) Since the sequence can operate as a four-level system, it should be readily adapted to a variety of excitation devices as the output should continue as long as the population of the energy storage level is replaced;
- 2) Since the operating wavelength is generally longer than the photoionization and photodissociation thresholds for all of the important species in the kinetic sequence pumping the inversion, operation at an arbitrarily high power density should be possible with no loss of efficiency;

and 3) Because the laser transition occurs between bound electronic states of a molecular ion, the bandwidth of the gain is small, and, hence, the maximum gain is large. As a consequence, saturation should be reached at smaller inversion densities than in excimer transitions.

The prototype of such charge transfer systems is the helium-nitrogen laser demonstrated by Collins, et.al.,⁶ in 1974. When excited in a plasma produced by the discharge of an intense electron

beam⁷ it exhibited all of these advantages specific to this laser type. With e-beam currents of the order of 1 KA/cm² quasi-cw operation of the laser was achieved⁴ at 427.8 nm and output power was found to accurately follow input power after the onset of threshold. A time-independent power efficiency of 3% was found and tended to confirm the operation of the laser as a four-level system.⁸ As a result, no evidence of bottlenecking was found over a range of circulating intracavity intensities up to 30 MW/cm². Peak power densities as great as 320 MW/l showed no evidence of disturbing the kinetic sequence pumping the laser transition.⁹

Early reports^{10,11,12} of the discharge excitation of the laser transition at lower current densities presented an efficiency lowered to 0.05% of the total stored energy. Other differences were found in the timing of the development of the laser pulse. While the results for e-beam excitation^{4,7,13} had indicated a delay of 5-10 nsec between the onset of the excitation current and the initiation of the laser output, a more immediate development of the laser output was observed from these discharge plasmas having greater gain pathlengths. Less than 1 nsec delay was observed with an output pulse risetime of the order of 2 nsec. Using the accepted values

of the binary rate coefficients, the lifetime against charge transfer from helium to nitrogen should have been in excess of 10 nsec at 3 atm. pressure of the gas compositions generally used. This apparent inability of charge transfer to pump the laser transition fast enough together with the lower efficiencies led early authors¹⁰ to misidentify the principal kinetic step as being direct collisional excitation of neutral nitrogen by hot electrons in the tail of the energy distribution and generally contributed some doubt in later work as to the similarity of the pumping mechanisms under the different conditions of excitation. However, the new multibody charge transfer reactions reported recently^{2,14} now resolve this discrepancy by providing the necessarily high rates of reaction. The relevant reaction of importance in the helium-nitrogen discharge laser is termolecular charge transfer from He_2^+ to N_2 , occurring with a rate coefficient of $1.6 \times 10^{-29} \text{ cm}^6 \text{ sec}^{-1}$. Table 1 summarizes the improvement in the expected speed of the reaction brought about by the inclusion of this step in the kinetic sequence. It is thus consistent with

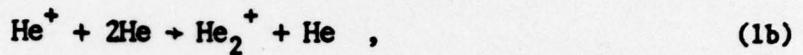
Table 1

Reaction times for charge transfer from He_2^+ as computed from traditional low pressure rate coefficients in comparison with values computed from recently obtained coefficients describing multibody processes. Gas composition is assumed to be 0.15% N_2 .

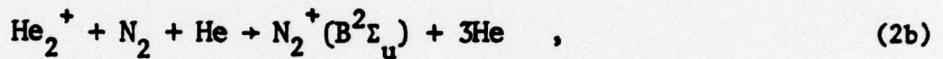
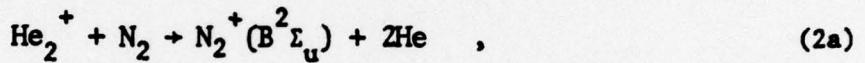
Pressure (atm)	Reaction Times	
	Bimolecular ¹ (nsec)	Multibody ² (nsec)
3	7.0	4.0
5	4.2	1.8

the timing of the laser outputs to attribute the operation of the helium-nitrogen laser under either e-beam, or direct discharge pumping to the common excitation scheme:

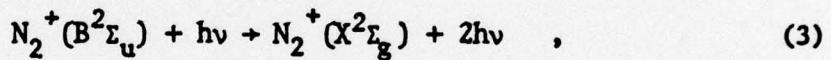
Ionization



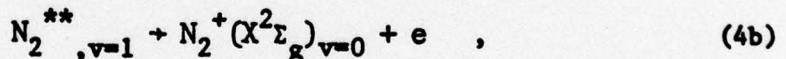
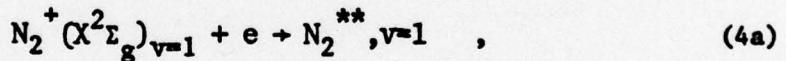
Charge Transfer



Stimulated Emission



Capture-Autoionization



where the double asterisk indicates an autoionizing level. The rate limiting step in the kinetic sequence is the charge transfer step.

The work reported here concerns a re-examination of the performance of the helium-nitrogen laser excited directly in a preionized discharge. It will be shown that lowered output efficiencies generally occurred

as the result of a loss of coupling between the electrical load and driving circuits caused by the time-varying impedance of the laser tube. At the E/p values of the order of $5 \text{ Vcm}^{-1}\text{torr}^{-1}$ and the current densities of 100 A cm^{-2} employed in this work power efficiencies of 2% were found, with peak powers in excess of 1MW. Thus, both in timing and efficiency the performance of the discharge pumped laser was found to be consistent with the results of the e-beam excitation and, hence, to be consistent with the charge transfer sequence summarized above.

II. EXPERIMENTAL METHOD

A schematic representation of the main discharge circuit together with the principal diagnostic circuitry used in the work reported here is shown in Fig. 1. The laser tube consisted of a machined delrin pressure vessel containing 87 cm. long electrodes of 0.3 cm thickness separated by either 1.3 cm or 1.7 cm. The current flow in the tube was transverse to the optical axis giving computed inductances for the laser tube of 0.23 and 0.3nH, respectively, for the two possible electrode spacings. Measured values agreed with the computed values to within experimental error. As shown in Fig. 1 the laser tube was connected in a Blumlein circuit switched by an EG&G 3202 hydrogen thyratron. The Blumlein capacitors marked C in the figure were constructed from 0.08 cm thick G-30 printed circuit board, copper clad on one side and contacted to two sheets of 0.039 cm thick mylar on the other. A foil electrode contacted to the outer surface of the mylar completed the capacitors giving a

value of $C=12nF$ for each.

In operation the electrical performance of the main discharge circuit resembled that of the neutral N_2 laser described and analyzed in detail by Fitzsimmons.¹⁵ Because of the relatively high inductance of the thyratron, the switching circuit functioned as a lumped LCR circuit which, upon commutation of the thyratron, tended to invert the voltage across the left capacitor in Fig. 1. The ringing period of the switching circuit was of the order of 100 nsec in this arrangement.

As the voltage across that capacitor proceeded toward a full inversion the voltage across the laser tube tended toward twice the original charging voltage. At some point, depending upon the pressure and gas composition in the laser tube, the E/p would reach a value sufficient to support the avalanche growth of the ion concentration in the gas mixture, thus initiating the kinetic chain pumping the laser transition. As the ionization increased, the conductivity increased permitting the Blumlein to discharge through the laser tube on a time scale short compared to the original ringing period of the switching circuit. In the atmospheric electrical avalanche (AEA) device finally realized in these experiments, discharge currents up to 30 KA could be attained with risetimes of a few nanoseconds. Switched voltages approaching 50 KV could be developed across the laser tube prior to breakdown.

In operation it was found that adequate preionization was necessary before the main discharge could develop uniform and stable plasmas. In the AEA laser developed for this work spatial uniformity

was finally achieved up to 9.3 atm. pressure in discharges with transverse aspect ratios varying from 1 x 1 to 6 x 1 through the use of displacement current preionization, a technique superior to UV-preionization in relatively "transparent" gases such as helium. A potential of the order of 40 to 50KV was applied to an electrode outside the pressure vessel of the laser tube in a manner to create an intense electric field perpendicular to the axis of discharge current flow and perpendicular to the optical axis. The high voltage pulse was developed by a cable transformer¹⁶ driven by a grounded grid thyratron switching a low-inductance capacitor. A delay of the order of 0.5 to 1.0 microsecond between the preionization pulse and the main discharge was found to be necessary. Since both the preionization and the main discharge were switched by hydrogen thyratrons the AEA laser could be readily operated at repetition rates from 1 to 30 Hz. Because of the longitudinal gas flow, excessive heating prevented the operation of the device at higher repetition rates. It appears that a transverse gas flow would allow operation at much higher repetition rates.

III. RESULTS

As shown in Fig. 1, the electrical performance was monitored by voltage and current probes connected to the laser tube. A tapped water resistor placed across the tube and inductively decoupled from the ground of a Tektronix 519 oscilloscope provided the means for measuring the voltage difference applied across the laser tube. The current flowing in the discharge loop was monitored with a $\frac{1}{2}$ loop

which could be rotated through 180° . In fact, this rotation proved necessary because of the capacitive coupling between the loop and the Blumlein. Each measurement of current was determined from the difference of two measurements of δ made with the plane of the loop being rotated through 180° .

Figure 2 shows typical data obtained from the voltage probe and illustrates the interaction of the Blumlein with the switching circuit. The voltage developed across the laser tube is shown for several values of pressure. In each case the charge voltage on the line was 24KV and the electrode spacing was 1.7cm. Shown by the dashed line is the open circuit ringing voltage obtained at values of pressure and preionization voltage sufficiently extreme that no discharge occurred. It agreed well with the simple model for the series LCR circuit for a switch inductance of 15nH, a value in good agreement with the thyratron specifications. As can be seen, the peak voltage across the laser tube was found to ring to a maximum of only 1.5 times the charge voltage instead of the doubling expected for an ideal switch. The solid curves represent typical operating conditions for the pressures in atmospheres indicated in Fig. 2. It can be seen that the loading of the switching circuit caused by the conductivity of the preionized gas caused a further reduction in the voltage developed across the tube prior to the onset of the electrical avalanche. In most cases it was found that with proper preionization the voltage actually appearing across the laser tube was roughly equal to the original charging voltage. As was expected, increases in pressure in the laser tube caused progressive increases

in the delay of the onset of the avalanche with corresponding increases in the voltage developed. In Fig. 2 the occurrence of the electrical avalanche can be seen to cause a rapid decrease of the voltage across the tube as the conductivity of the gas increased to meet the inverse impedance of the Blumlein driving circuit. Optimal performance of the laser was found to correspond to conditions for which the duration of the avalanche spanned the maximum in the open circuit voltage curve. For example, for a charging voltage of 24KV, Fig. 2 indicates that the optimal performance would occur for pressures of the order of 3.7 atm in agreement with the maximum observed in the laser output. Conversely, at 5.1 atm the developing avalanche can be seen to be competing for stored charge with the "ringing down" of the voltage in the switching circuit. A correspondingly lower laser output was observed in that case.

Figure 3 shows a comparison of typical data obtained from the voltage and current diagnostic devices. Shown by the dashed curve is the difference obtained between successive measurements made with the B loop after rotation through 180° . It was observed that the data obtained with the two orientations generally showed the proper inversion but contained an error signal which did not change sign and which represented about 20% of the amplitude. Computing the difference of the data obtained for the two orientations effectively removed this error signal and led to the type of data for dI/dt shown in the figure.

It can be seen in Fig. 3 that the successive nodes of the dI/dt curve correspond to the nodes of the direct measurement of the voltage across the load as plotted by the dotted curve. At these

nodes, since $dI/dt=0$, the voltage must represent only that component appearing across the resistive part of the load, namely the plasma. Since at that point that voltage is also zero, it can be concluded that by that time the avalanche has proceeded to an end-point at which the resistance of the load has fallen to a value comparable to the Blumlein impedance and no further increase in ionization can occur. At later times the voltage excursions seen in Fig. 3 must represent only the voltage developed across the inductive part of the laser tube as the current continued to ring in the Blumlein circuit. A comparison of the relative amplitudes of the oscillations of the voltage curve and of the dI/dt curve showed them to be consistent with the calculated value of tube inductance. Then, for subsequent analytical purposes the measured voltage was corrected by multiplying the value of tube inductance by the measured value of dI/dt and subtracting it from the measured voltage. A typical result for the voltage appearing across resistive part of the load and, thus, leading to dissipation is shown in Fig. 3 by the heavy solid curve. The time dependent current obtained by integrating the dI/dt curve is shown for comparison by the light solid curve. The instantaneous power dissipation in the load can be obtained by forming the product of the two curves.

A comparison of the time-dependence of the input power to the laser output power showed that the smaller electrode spacing resulted in a better power conversion, probably because of a more thorough saturation of the optical transition. Results for the 1.3 cm spacing of the electrodes are shown in Fig. 4 for the four values of gas pressure indicated, each containing 0.15% N_2 . The charge voltage was 24KV and the repetition rate was 10Hz. Dashed lines show the

discharge currents and the voltages appearing across the resistive part of the load. The powers dissipated in the load are shown by the dotted curves and the solid curves record the laser output power measured in the (0+1) vibrational component of the B+X electronic transition of N₂ at 427.8 nm.

The pressure-dependent delays between the input powers applied to the plasma and the laser outputs are consistent with the magnitudes of the reaction times for the charge transfer step as shown in Table 1. The scales for the power have been chosen as indicated, so that the close correlation between input and output power on the leading edge of the curves for the high pressure plasmas corresponds to an instantaneous conversion efficiency of 2%. Integrals under the power curves shown in Fig. 4 give values for the energies of the laser pulses of 1.3, 2, 3, and 4mJ, respectively for the pressures varying from 3 to 5.1 atm. These results in comparison with the integrals under the input power curves yield efficiencies of 1.3, 1.0, 1.0, and 0.8% respectively, for the conversion of the energies dissipated in the load for the cases shown in Fig. 4.

The field strengths in the plasma corresponding to the maximum observed transfer of power can be determined also from Fig. 4. At the higher pressures where the delay in the kinetic chain is minimal, the highest instantaneous power efficiency can be seen to occur on the leading edge of the pulses where the E/p is greatest. For the times at which 2% efficiency was sustained the E/p can be computed from

Fig. 4 to have been decreasing from 3.5 to $2 \text{ V cm}^{-1}\text{torr}^{-1}$. The corresponding current densities were increasing from zero to 160A cm^{-2} . It can be seen that the loss of efficiencies for the conversion of the pulse energy at the higher pressures resulted from the prolonged flow of discharge current at relatively low E/p .

IV. CONCLUSIONS

The parameterization of the charge transfer AEA laser is summarized in Fig. 5 for the case of the 1.7 cm electrode spacing. The peak laser power observed has been plotted as a function of gas pressure and parametrically as functions of charging voltage on the Blumlein. The gross behavior is evidently dominated by the degree of the coupling of the plasma load to the electrical driving circuit. A comparison of the curve corresponding to 24KV charging voltage with the data of Fig. 2 shows that the peak output occurred for the case in which the avalanche developed and reached completion during the period the switching circuit was ringing through the maximum in voltage as discussed previously. Computed in terms of the maximum switched voltage the peaks in the performance curves of Fig. 5 correspond approximately to an E/p value of $5\text{V cm}^{-1}\text{torr}^{-1}$.

In conclusion it appears that the AEA discharge device represents a useful means for exciting the helium-nitrogen charge transfer laser and peak powers reaching 1 MW have been demonstrated. Instantaneous power transfer efficiencies of 2% have been achieved at E/p values of the order of $3 \text{ V cm}^{-1}\text{torr}^{-1}$ and current densities of 100 to 200A cm^{-2} .

The corresponding values of efficiency for the conversion of energies dissipated in the laser tube into output pulse energies have been shown to be around 1%.

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5. This process is the analog for molecular ions to the rapid vibrational quenching of neutral molecules by the capture of free electrons to form unstable negative ions. However, since the interaction force is Coulombic for the molecular ions in comparison with the weaker charge-induced dipole forces for the neutrals, the quenching of vibration by electron collisions should proceed even more rapidly in the case of the molecular ions. For a detailed discussion of this process see J. Stevafelt, *Phys. Rev. A8*, 2507 (1973).
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CAPTIONS

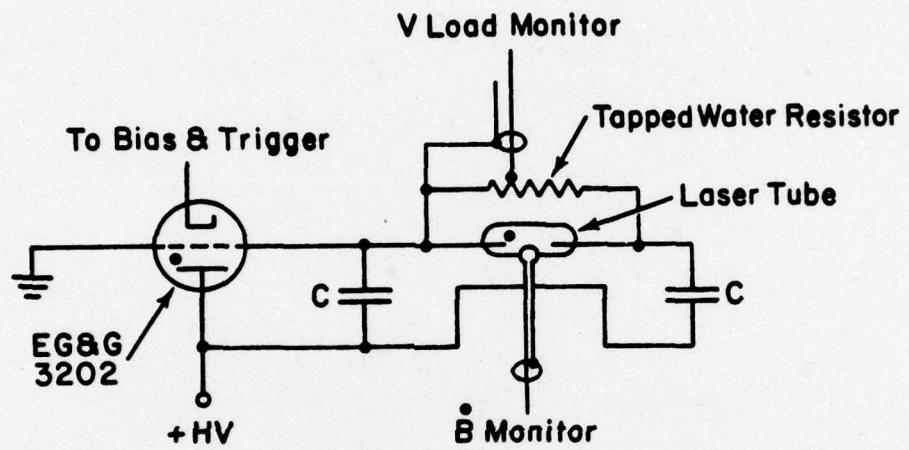
Figure 1: Schematic diagram of the circuit of the Atmospheric Electrical Avalanche (AEA) Laser used in these experiments.

Figure 2: Graphs of the transient voltage switched across the laser tube. The dashed curve shows the open circuit voltage ringing across the laser tube when the electrical breakdown is inhibited. Solid curves record the voltage appearing across the laser tube when filled to the indicated pressure in atmospheres and preionized. The electrode spacing was 1.7 cm.

Figure 3: Graphs of typical diagnostic data obtained from the \dot{B} loop and from the voltage divider connected across the laser tube are shown by the dashed and dotted curves, respectively. The light solid curve shows the current obtained by integrating the dashed curve and the heavy solid curve records the voltage appearing across the resistive part of the load obtained by removing the inductive component from the dotted curve.

Figure 4: Graphs comparing the electrical and optical performance of the laser tube obtained for a 1.3 cm electrode spacing and for the indicated pressures of helium containing 0.15% N₂. Dashed curves plot the voltage and current applied to the laser tube as marked and scaled to the leftmost ordinate. The dotted curve records the instantaneous power dissipation in the laser tube and is scaled to the middle ordinate. The solid curve plots the output power of the laser measured at 427.8 nm and is scaled to the rightmost ordinate.

Figure 5: Graph showing the parameterization of the charge transfer AEA laser operating with 1.7 cm electrode spacing. Peak output power is shown as a function of helium pressure and parametrically as functions of the charge voltages shown.



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